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This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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TITLE OF THE INVENTION (280 characters max)

LOW ACTIVATION ADHESIVE COMPOSITION CONTAINING POLYOLEFIN/FUNCTIONAL POLYMERS/ HIGH IMPACT POLYSTYENE WITH HIGH PEEL STRENGTH AND COHESIVE FAILURE MODE

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LOW ACTIVATION ADHESIVE COMPOSITION CONTAINING POLYOLEFIN/FUNCTIONAL POLYMERS/ HIGH IMPACT POLYSTYENE WITH HIGH PEEL STRENGTH AND COHESIVE FAILURE MODE

This invention relates to new adhesive compositions, and in particular to adhesives that can be applied as a self-supporting film, co-extruded or extrusion coated on a substrate.

The adhesive composition described herein is a melt blend of number of polymers. Such adhesives could be used in all kinds of applications and possess varying properties depending on the application. One application for this adhesive is in manufacturing metal panels for the construction industry in which the panels are a composite structure where one side is a metal substrate such as aluminum or steel and the other side is a polyolefin such as polyethylene. The adhesive is used to bond the polyolefin to the metal substrate.

In such an application, it is important for the adhesive layer to provide excellent adhesion to polyolefin and to the metal substrate. As well, it is highly desired that the adhesive fail cohesively during testing and should be relatively easy to formulate and use. The adhesive compositions of the present invention provide adhesives with excellent adhesion to metalic substrates as well as a number of polymeric materials.

The adhesives of the present invention provide an excellent balance of high peel force and cohesive mode of failure, which exceed that of adhesive compositions currently used in the marketplace. Moreover, the adhesive compositions of the present invention activate at lower temperatures than adhesives currently used in aluminum building panels. This lowering of the lamination temperature results in considerable cost savings and safer operations for panel manufacturers because the use of protective layers can be eliminated from the manufacturing process.

Accordingly, the present invention provides adhesive compositions that possess excellent bonding properties to metallic substrates and to a number of polymeric materials resulting in an adhesive layer with high peel strength and cohesive mode of failure. The present invention also allows for a low activation temperature to be used in the manufacture of building panels.

The adhesive compositions of the present invention may be marketed as an

adhesive resin in pellet form or as an adhesive film.

In its broadest aspects, the adhesive composition of the present invention comprises:

a) 10 to 90 wt% polyolefin;

b) 5 to 90 wt% of a grafted polyolefin;

c) 5 to 40 wt% high impact polystyrene; and

d) 0 to 30 wt% of a rubber compound selected from ethylene-propylene diene and polyisobuthylene.

"Polyolefin" means homopolymers and copolymers of unsaturated hydrocarbons having 2-20 carbon atoms. They can be made by processes well known in the art, including metallocene processes. In particular, the polymers are homopolymers of ethylene or propylene or copolymers of ethylene with one or more alpha-olefin hydrocarbons having 3-10 carbon atoms, especially propylene, butene-1, hexene-1 and octene-1 and styrene. Suitable alpha-olefins also include dienes, that is, monomers with more than 1 site of unsaturation, especially 1,3 butadiene, 1,5 hexadiene and norbornadiene. In some embodiments, the Polyolefins are copolymers of ethylene with a hydrocarbon alpha-olefin having from 4-8 carbon atoms. Mixtures and blends of the Polyolefins may be used.

The term "grafted polyolefin" refers to a polyolefin on which is grafted at least one monomer selected from ethylenically unsaturated carboxylic acids and ethylenically unsaturated carboxylic acid anhydrides, including less preferably, derivatives of such acids, and mixtures thereof. Examples of the acids and anhydrides, which may be mono-, di- or polycarboxylic acids are acrylic acids, methacrylic acid, maleic acid, fumaric acid, itatonic acid, crotonic acid, itatonic anhydride, maleic anhydride and substituted maleic anhydride, e.g. dimethyl maleic anhydride or citrotonic anhydride, nadic anhydride, nadic methyl anhydride, and tetrahydrophthalic anhydride, maleic anhydride being particularly preferred. Examples of the derivatives of the unsaturated acids are salts, amides, imides and esters, e.g., mono- and disodium maleate, acrylamide, glycidyl methacrylate and dimethyl fumarate.

In one embodiment of the present invention, an adhesive composition is provided that comprises the following components:

(a) 10 to 90 wt%, preferably 40 to 60 wt%, and most preferably 44.9 wt% of ethylene vinyl acetate copolymer (for example, commercially available as Elvax 3130 available from DuPont);

(b) 5 to 90 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of maleic anhydride grafted polyethylene (for example, commercially available as Fusabond E MB-439D from DuPont);

(c) 5 to 40 wt%, preferably 10 to 20 wt%, and most preferably 15.0 wt% of high impact polystyrene (for example, commercially available as HCC850 from Nova Chemicals);

(d) 0 to 30 wt%, preferably 10 to 20 wt%, and most preferably 15.0 wt% of an ethylene-propylene diene rubber compound (for example, commercially available as Nordel IP 3720 from DuPont-Dow Elastomers); and

(e) 0.1 to 0.5 wt%, preferably 0.1 wt% of an antioxidant (for example, commercially available as ANOX 20N from Ciba-Geigy).

In a second embodiment of the present invention, an adhesive composition is provided that comprises the following components:

(a) 10 to 90 wt%, preferably 40 to 60 wt%, and most preferably 29.9 wt% of metallocene linear low density polyethylene (for example, commercially available as Exact 3024 from Exxon);

(b) 5 to 90 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of maleic anhydride grafted polyethylene (for example, commercially available as Fusabond E MB-265D from DuPont);

(c) 5 to 40 wt%, preferably 25 to 35 wt%, and most preferably 30.0 wt% of high impact polystyrene (for example, commercially available as HCC850 from Nova Chemicals);

(d) 0 to 30 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of an ethylene-propylene diene rubber compound (for example, commercially available as Nordel IP 3720 from DuPont-Dow Elastomers); and

(e) 0.1 to 0.5 wt%, preferably 0.1 wt% of an antioxidant (for example, commercially available as ANOX 20N from Ciba-Geigy).

In these two embodiments, polyisobuthylene may be used instead of the ethylene-

propylene diene rubber compound. As well, high impact polystyrene (styrene-butadiene rubber) having different butadiene rubbers (from 4.25-9.5wt%) may be used with similar effects.

Each component of the adhesive composition blend provides certain attributes to the final adhesive. The low melting point polyolefin, such as polyethylene or ethylene vinyl acetate copolymer provides the low activation temperature property of the adhesive. The functional polymer mainly contributes to the superior bonding of the adhesive composition to metal substrates, whereas the rubber component toughens the final adhesive compositions. Finally, the high impact polystyrene component provides the adhesive with superior cohesive failure properties and also contributes to improved toughness.

The adhesive compositions of the present invention have melt flow rates of 1 to 5 dg/min. (ASTM-1238E). The melting point is between 80°C- 140°C depending on the composition.

Because of their combination of high peel strength, cohesive failure mode and bonding at low temperature, the adhesives of the present invention are useful in the manufacture of aluminum and steel building panels.

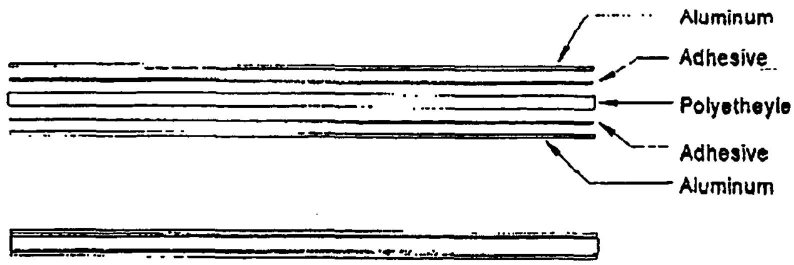
Examples

The following examples show the advantages of using High Impact Polystyrene in providing additional peel strength, cohesive mode of failure and a non-tacky adhesive film.

Example 1:

Table 1 shows the compositions A1, A2 and A3 at 0 wt%, 10 wt% and 20 wt% High Impact Polystyrene ("HIPS") respectively. These compositions were dry-blended initially and fed to a small co-rotating twin screw extruder. The melt compounded materials were pelletized and then blown into film with an average thickness of about 75 microns. The films were subsequently used as adhesive layers to make a 5 layer composite structure as shown in Figure 1. The aluminum was aludyne treated and had a thickness of 0.38 mm. Polyethylene core used was low density polyethylene (LDPE) sheet, 2 mm thick.

Figure 1: 5-Layer Laminated Structure



The lamination of the structures were carried out in a electrically heated press as follows:

1- Preheat the 5 layer structure for 4.5 min. @ 100 °C.

2- Apply pressure of 5 kgf/cm^2 at 135 °C for 20 seconds.

3- Release the pressure and leave the composite in the press for an additional 1.5 min. @ 135°C.

4- Air cool the sample to room temperature.

The composite structure was then tested to measure the peel strength (ASTM 1876) using an Instron™ machine as shown in Figure 2. The following conditions were used during the peel strength test:

i) Cross-head speed: 100 mm/min.

ii) Peel mode: 180° T-peel

Figure 2: 180° T-Peel test of Laminated Structure

The last two columns in Table 1 indicate the results of the peel strength and failure mode as the amount of Component 4 (HIPS) is increased from 0 wt% to 20 wt%. It is also highly desirable to achieve a cohesive failure (50-50 to polyethylene and aluminium). Sample A3 shows 80% cohesive failure as compared to adhesive failure in the other two compositions.

Table 1: Ethylene Vinyl Acetate (EVA) based Adhesives:

[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
A1	Elvax 3170 49.9%	Fusabond E- MB 439D 30.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 0.0%	Anox 20N 0.1%	16.4	Adhesive to Al.
A2	Elvax 3170 39.9%	Fusabond E- MB 439D 30.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 10.0%	Anox 20N 0.1%	23.6	Adhesive to Al.
A3	Elvax 3170 29.9%	Fusabond E- MB 439D 30.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.1%	29.8	80% Cohesive
D1	Elvax 3170 29.9%	Fusabond E- MB 439D 30.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.1%	16.5	100% Cohesive
D6	Elvax 3130 44.9%	Fusabond E- MB 439D 20.0%	Nordel IP-3720 15.0%	HIPS (HCC850) 20.0%	Anox 20N 0.1%	17.5	100% Cohesive
D7	Elvax 3130 34.9%	Fusabond E- MB 439D 20.0%	Nordel IP-3720 15.0%	HIPS (HCC850) 30.0%	Anox 20N 0.1%	10.9	100% Cohesive

Example 2:

Samples were prepared as described in Example 1 with the exception that co-extruded blown films of polyethylene/Adhesive were made and substituted for a pure adhesive film in order to reduce the cost of the adhesive used. The compositions D1, D6 and D7 are co-extruded LLDPE-Adhesive (25 micron each). The co-extruded films generally result in significantly lower peel strength as can be seen compering A3 vs. D1 (29.8 vs. 16.5 kgf/25mm, respectively). Composition d6 appeared to be optimal in peel strength. Further addition of High Impact Polystyrene from 20 wt% to 30 wt% significantly reduced the peel strength (17.5 and 10.9 kgf/25 mm, respectively).

Example 3:

In this example, compositions were made using metallocene linear low density polyethylene as the base resin. Table II summarizes the compositions. Samples B1, B8 and B16 were prepared as those described in Examples 1 and 2. The last two compositions E1 and E2 were prepared in a 2-step process in order to more closely simulate the actual commercial production. Adhesives were initially laminated to aluminium at a temperature of 149 °C, and pressure of 8.5 kgf/cm^2 for 30 seconds. These pre-laminated aluminium sheets were then pressed with 93°C pre-heated LDPE core at 132°C, pressure of 8.5 kgf/cm^2 for 10 seconds. The peel testing was at 90° angle as opposed to the 180° peel test used in the other examples. Composition E1 is a repeat of B16 used to compare the new lamination method. It can be seen that composition E2 results in higher peel strength and 100% cohesive failure. It should be noted that competitor grades currently used in commercial production totally fail when using this "low lamination temperature" of 132°C.

Table 2: Metalocene LLDPE based Adhesives:

[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
B1	Exact 3024 59.9%	Fusabond E- MB 265D 20.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 10.0%	Anox 20N 0.10%	N/A	Adh. To Al.
B8	Exact 3024 49.9%	Fusabond E- MB 265D 20.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.10%	26.9	Adh. To Al.
B16	Exact 3024 39.9%	Fusabond E- MB 265D 20.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.10%	36.3	20% Cohesive
E1	Exact 3024 39.9%	Fusabond E- MB 265D 20.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.10%	18.6	30% Cohesive
E2	Exact 3024 29.9%	Fusabond E- MB 265D 20.0%	Nordel IP-3720 20.0%	HIPS (HCC850) 20.0%	Anox 20N 0.10%	20.5	100% Cohesive

I Claim:

1. An adhesive composition comprising:
   (a) 10 to 90 wt% polyolefin;
   (b) 5 to 90 wt% of a grafted polyolefin;
   (c) 5 to 40 wt% high impact polystyrene; and
   (d) 0 to 30 wt% of a rubber compound selected from ethylene-propylene diene and polyisobuthylene.

2. An adhesive composition comprising:
   (a) 10 to 90 wt%, preferably 40 to 60 wt%, and most preferably 44.9 wt% of ethylene vinyl acetate copolymer;
   (b) 5 to 90 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of maleic anhydride grafted polyethylene;
   (c) 5 to 40 wt%, preferably 10 to 20 wt%, and most preferably 15.0 wt% of high impact polystyrene;
   (d) 0 to 30 wt%, preferably 10 to 20 wt%, and most preferably 15.0 wt% of an ethylene-propylene diene rubber compound; and
   (e) optionally, 0.1 to 0.5 wt%, preferably 0.1 wt% of an antioxidant.

3. An adhesive composition comprising:
   (a) 10 to 90 wt%, preferably 40 to 60 wt%, and most preferably 44.9 wt% of metallocene linear low density polyethylene;
   (b) 5 to 90 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of maleic anhydride grafted polyethylene;
   (c) 5 to 40 wt%, preferably 25 to 35 wt%, and most preferably 30.0 wt% of high impact polystyrene;
   (d) 0 to 30 wt%, preferably 15 to 25 wt%, and most preferably 20.0 wt% of an ethylene-propylene diene rubber compound; and
   (e) optionally, 0.1 to 0.5 wt%, preferably 0.1 wt% of an antioxidant.

4. A composite structure comprising:

(a) a metal substrate;

(b) a polymeric layer; and

(c) a layer of the adhesive composition of claims 1, 2 or 3 between the metal substrate and the polymeric layer.